


Dipole resonances of nonabsorbing dielectric nanospheres in the optical range: Approximate explicit conditions for high- and moderate-refractive-index materials

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In this work, we discuss the way in which electric and magnetic dipole resonances arising in the optical scattering spectrum of nonabsorbing dielectric nanospheres can be accurately approximated by means of simple explicit expressions that depend on the sphere's radius, incident wavelength, and relative refractive index. We find such expressions to hold not only for high- but also for moderate-refractive-index values, thus complementing the results reported in previous studies.

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I. INTRODUCTION

Scattering of light by metallic nanoparticles shows a strongly resonant behavior within the optical region, which permits us to consider them as a sort of optical antennas [1], given their ability to redirect freely propagating light into localized energy, and vice versa. When in the subwavelength regime, such resonant nanoparticles may even be used as building blocks for optical metamaterials [2,3] or metaobjects [4–6]. Although systems based on metallic nanoparticles have raised the prospect of some very promising applications [7,8], they also suffer from two significant limitations when operated within the optical range: they are intrinsically lossy and do not exhibit any intrinsic magnetic response. As a consequence of this, many efforts have been recently devoted to obtain the same functionalities by means of nonabsorbing purely dielectric nanoparticles [9–14], which show magnetic resonances arising from the circulation of light-induced internal displacement currents.

From all possible dielectric nanoparticles, spherical ones are especially well suited to be used as nanoresonators, given their ease of synthesis by either chemical or physical methods and the fact that Mie theory [15] explicitly provides the scattering efficiency of a sphere as a function of the incident wavelength, the sphere's radius, and its relative refractive index with respect to that of the surrounding medium. Hence, different arrangements have been proposed for purposes of sensing [16,17] and directional control of scattered radiation [18–22] that are based on the selective excitation of resonances at dielectric nanospheres. In most of these proposals, the obtained scattering response is mainly dominated by dipole resonances, which are those with the lowest energy.

If it were possible to predict the occurrence of a dipole resonance for a triplet of sphere's radius, incident wavelength, and refractive index value without the actual evaluation of Mie scattering coefficients, this would undoubtedly result in

the easing of nanosphere-based designing. Some previous studies have partially achieved such an objective: Explicit expressions for resonances with any multipolar order and any ordinal number arising in nonabsorbing high-refractive index spheres have been presented in a recent paper [23]. Other authors have obtained similar results for the resonances with the lowest ordinal number of every multipolar order that can be excited in a sphere with a generic real [24] or complex refractive index [25,26]. In this work, we discuss the way in which triplets giving rise to electric and magnetic dipole resonances with any ordinal number in nonabsorbing spheres can be approximately determined from simple explicit expressions that hold not only for high-refractive-index but also for moderate-refractive-index values, thus complementing those reported in the above-mentioned references.

The paper is structured as follows: In Sec. II we review the basics of light scattering by a nonabsorbing dielectric sphere and introduce scattering coefficients and related magnitudes. Section III details the way in which dipole resonances arising in the scattering efficiency of high- and moderate-refractive-index spheres can be accurately approximated without the need for full Mie calculation. The validity of these approximations within the optical range for spheres made of Si, Cu₂O, and TiO₂ is discussed in Sec. IV. Finally, in Sec. V we summarize our work.

II. SCATTERING OF LIGHT BY A NONABSORBING DIELECTRIC SPHERE

Let us suppose that a uniform, nonmagnetic, and nonabsorbing dielectric sphere with radius R is surrounded by an also nonmagnetic and nonabsorbing dielectric medium. The dimensionless scattering efficiency for light propagating through the surrounding medium with wavelength λ can then be expressed as

$$Q_{\text{sca}}(x, m) = \frac{2}{x^2} \sum_{l=1}^{\infty} (2l+1) [|a_l(x, m)|^2 + |b_l(x, m)|^2], \quad (1)$$

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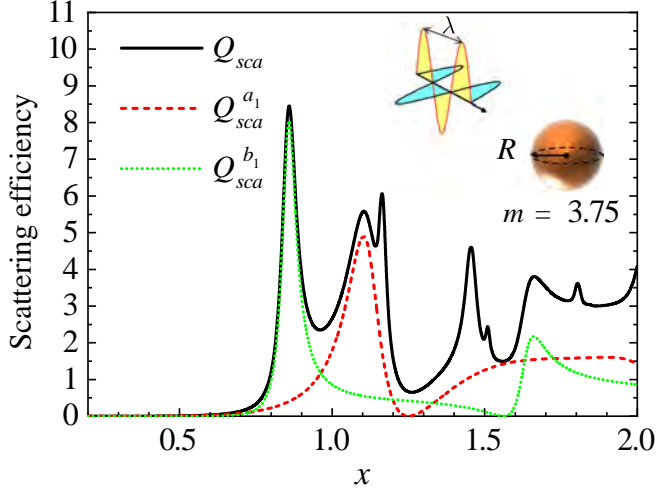


FIG. 1. Calculated scattering efficiency Q_{sca} (solid line) as a function of size parameter x for a dielectric sphere with $m = 3.75$. Dashed and dotted curves show the values of $Q_{\text{sca}}^{a_1}$ and $Q_{\text{sca}}^{b_1}$, respectively.

where $x = 2\pi R/\lambda > 0$ is the size parameter and m the relative refractive index of the sphere with respect to that of the medium [27]. The dependence of Q_{sca} on m (and mostly on x) is contained in the scattering coefficients a_l and b_l , which represent, respectively, the subsequent electric and magnetic contributions to the multipolar expansion (that is, dipole for $l = 1$, quadrupole for $l = 2$, octupole for $l = 3$, hexadecapole for $l = 4$, ...) of scattered fields.

Following Refs. [28,29], we find it convenient to write a_l, b_l in the form

$$a_l(x, m) = \frac{p_l(x, m)}{p_l(x, m) + iq_l(x, m)}, \quad (2a)$$

$$b_l(x, m) = \frac{r_l(x, m)}{r_l(x, m) + is_l(x, m)}, \quad (2b)$$

where

$$p_l(x, m) = m\psi_l(mx)\psi'_l(x) - \psi_l(x)\psi'_l(mx), \quad (3a)$$

$$q_l(x, m) = -m\chi'_l(x)\psi_l(mx) + \chi_l(x)\psi'_l(mx), \quad (3b)$$

$$r_l(x, m) = m\psi_l(x)\psi'_l(mx) - \psi_l(mx)\psi'_l(x), \quad (3c)$$

$$s_l(x, m) = -m\chi_l(x)\psi'_l(mx) + \chi'_l(x)\psi_l(mx). \quad (3d)$$

In Eqs. (3), $\psi_l(z) = zj_l(z)$ and $\chi_l(x) = -xy_l(x)$ are the Ricatti-Bessel functions, which are connected, respectively, to the spherical Bessel functions j_l and y_l [30]. The prime denotes the derivative with respect to the entire argument of the corresponding function. Please notice that the convenience of writing the scattering coefficients in this fashion rests on the fact that auxiliary functions p_l, q_l, r_l , and s_l can only take real values as a direct consequence of m 's being real [31]. This also prevents divergencies in Q_{sca} , which shows resonances if either $q_l(x, m)$ or $s_l(x, m)$ vanish. Given m and l , there are infinitely many positive values of x that fulfill such conditions, due to the oscillatory nature of the Ricatti-Bessel functions.

As an illustration of this behavior, we present in Fig. 1 the calculated scattering efficiency Q_{sca} (solid line) as a function of size parameter x for a sphere with $m = 3.75$, which corresponds to the average value of the range $m = 2.5$ to 5 . In order to point out the origin of different resonances, we also include the specific contribution of dipole terms by means of auxiliary quantities $Q_{\text{sca}}^{a_1} = 6|a_1|^2/x^2$ (dashed line) and $Q_{\text{sca}}^{b_1} = 6|b_1|^2/x^2$ (dotted line). As can be seen, dipole contributions dominate the scattering response for $x \lesssim 1.15$, with magnetic and electric resonances located at $x = 0.86$ and 1.10 , respectively.

Let us consider that the sphere is made by silicon and surrounded by air. Hence, the incident wavelength for its relative refractive index to be 3.75 is 720 nm [32]. This implies that a sphere with radius $R \approx 99$ nm will show a magnetic dipole resonance for that particular wavelength, which, in contrast, will give rise to a mostly electric resonance for $R \approx 126$ nm. If one increases the sphere's radius up to 190 nm (that is $x = 1.66$), $Q_{\text{sca}}^{b_1}$ will show another peak, although the total scattering efficiency significantly reduces with respect to its value for $x = 0.86$. It is then clear that, given two of the three m, λ , and R parameters, dipole resonances can only appear for some specific values of the third one. In the following sections, we will discuss the way in which resonant (m, λ, R) triplets can be approximately determined without the actual evaluation of neither a_1 nor b_1 .

III. APPROXIMATE DETERMINATION OF ELECTRIC AND MAGNETIC DIPOLE RESONANCES

On the assumption that m is kept as a constant, let $\{x_{\text{res}}^{a_1,1}, x_{\text{res}}^{a_1,2}, \dots, x_{\text{res}}^{a_1,j}, \dots\}$ be the set of infinitely many positive solutions to

$$m\chi'_1(x_{\text{res}}^{a_1,j})\psi_1(mx_{\text{res}}^{a_1,j}) = \chi_1(x_{\text{res}}^{a_1,j})\psi'_1(mx_{\text{res}}^{a_1,j}), \quad (4)$$

where $j = 1, 2, \dots$ is a positive integer number. Hence, an electric dipole resonance appears in Q_{sca} for any pair of values of R and λ that meet the condition $R/\lambda = x_{\text{res}}^{a_1,j}/2\pi$, with the caveat that m also depends on λ . For magnetic dipole resonances, we can then define $\{x_{\text{res}}^{b_1,1}, x_{\text{res}}^{b_1,2}, \dots, x_{\text{res}}^{b_1,j}, \dots\}$ as the analog infinite set of positive solutions to

$$m\chi_1(x_{\text{res}}^{b_1,j})\psi'_1(mx_{\text{res}}^{b_1,j}) = \chi'_1(x_{\text{res}}^{b_1,j})\psi_1(mx_{\text{res}}^{b_1,j}). \quad (5)$$

In order to determine $\{x_{\text{res}}^{a_1,j}\}$ and $\{x_{\text{res}}^{b_1,j}\}$, let us now take a closer look to the explicit form of the Ricatti-Bessel functions for $l = 1$:

$$\psi_1(mx) = \frac{\sin mx}{mx} - \cos mx, \quad (6a)$$

$$\psi'_1(mx) = -\frac{\sin mx}{(mx)^2} + \frac{\cos mx}{mx} + \sin mx, \quad (6b)$$

$$\chi_1(x) = \frac{\cos x}{x^2} + \sin x, \quad (6c)$$

$$\chi'_1(x) = \cos x - \frac{2 \cos x}{x^3} - \frac{\sin x}{x^2}. \quad (6d)$$

It is clearly apparent that Eqs. (6) can be greatly simplified for some limiting values of x and m , thus making it easier to

solve Eqs. (4) and (5). In particular, we will consider three different scenarios that are hereafter described in order of increasing complexity.

A. Approximations to $x_{\text{res}}^{a_1,j}$ and $x_{\text{res}}^{b_1,j}$ for $x \gtrsim 1$; $m \gg 1$

It can be shown from Eqs. (6) that both $|\chi_1 \psi'_1|$ and $|\chi'_1 \psi_1|$ are less than one for $x \gtrsim 1$ whatever the value of m . For $m \gg 1$, functions q_1 and s_1 can therefore be approximated as

$$q_1(x, m) \approx -m\chi'_1(x)\psi_1(mx), \quad (7a)$$

$$s_1(x, m) \approx -m\chi_1(x)\psi'_1(mx). \quad (7b)$$

As far as $mx \gg x$, solutions to Eqs. (4) and (5) will then be defined by the following conditions [23]:

$$\psi_1(mx_{\text{res}}^{a_1,j}) \approx 0, \quad (8)$$

$$\psi'_1(mx_{\text{res}}^{b_1,j}) \approx 0. \quad (9)$$

Let us now assume that mx is large enough to disregard all but purely sinusoidal terms in Eqs. (6a) and (6b). Hence, determination of resonances simplifies even more, as size parameters would only have to meet the conditions of Eqs. (8) and (9) up to the zeroth order in powers of $1/mx$:

$$\cos mx_{\text{res}(0)}^{a_1,j} = 0, \quad (10a)$$

$$\sin mx_{\text{res}(0)}^{b_1,j} = 0, \quad (10b)$$

the extra subscript being added in order to avoid any confusion with subsequent results hereafter.

From Eqs. (10), the well-known expressions

$$x_{\text{res}(0)}^{a_1,j}(m) = \frac{(j + \frac{1}{2})\pi}{m}, \quad (11)$$

$$x_{\text{res}(0)}^{b_1,j}(m) = \frac{j\pi}{m} \quad (12)$$

are readily obtained. With respect to Eq. (11), it has to be pointed out that we have set the zeroth-order fundamental resonance to $3\pi/2m$ (and not to $\pi/2m$) in order to be consistent with $x \gtrsim 1$. Nevertheless, we would rather preserve the full version of Eqs. (8) and (9) and find their solutions by expanding in a Taylor series about $x_{\text{res}(0)}^{a_1,j}$ and $x_{\text{res}(0)}^{b_1,j}$ [cf. Eqs. (8.13) and (8.14) in Ref. [23]]:

$$x_{\text{res}(1)}^{a_1,j}(m) = x_{\text{res}(0)}^{a_1,j}(m) \left[1 - \frac{1}{(j + \frac{1}{2})^2 \pi^2 - 1} \right], \quad (13)$$

$$x_{\text{res}(1)}^{b_1,j}(m) = x_{\text{res}(0)}^{b_1,j}(m) \left[1 - \frac{1}{j^2 \pi^2 - 2} \right]. \quad (14)$$

As far as the obtained expressions are nothing other than the sequential zeros of $j_1(mx)$ and $mxj_0(mx) - j_1(mx)$, their numerical precision can be extended on demand by means of standard techniques [30].

B. Improved approximations to $x_{\text{res}}^{a_1,1}$ and $x_{\text{res}}^{b_1,1}$

According to Eq. (13), we expect the size parameter for fundamental electric dipole resonance to be approximately equal to 1.43 in units of π/m . However, as can be seen

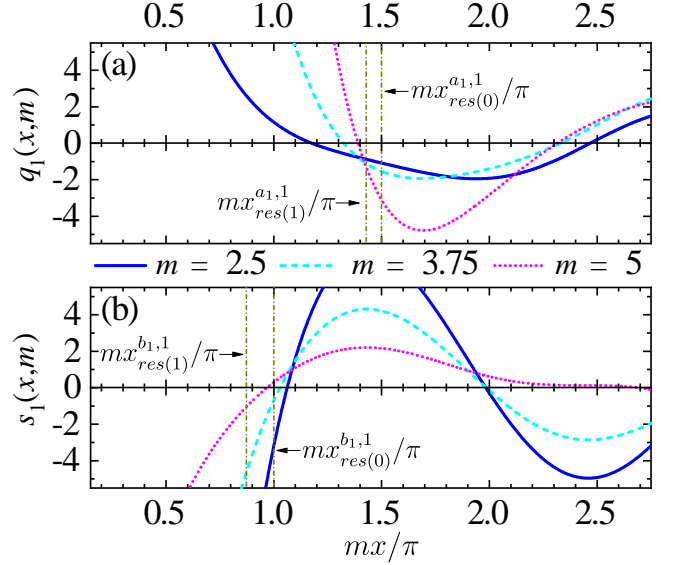


FIG. 2. Calculated $q_1(x, m)$ (a) and $s_1(x, m)$ (b) as a function of mx/π . Solid, dashed, and dotted curves show the values for $m = 2.5, 3.75$, and 5 , respectively. Vertical dashed-dotted lines mark the values of $x_{\text{res}(0)}^{a_1,1}$, $x_{\text{res}(1)}^{a_1,1}$, $x_{\text{res}(0)}^{b_1,1}$, and $x_{\text{res}(1)}^{b_1,1}$ in units of π/m .

in Fig. 2(a), that value actually defines some upper bound that is not reached even for $m = 5$. For the fundamental magnetic dipole resonance in Fig. 2(b), $x_{\text{res}(0)}^{b_1,1}$ seems to provide a better approximation than $x_{\text{res}(1)}^{b_1,1}$, although neither of them completely capture the dependence of $x_{\text{res}}^{b_1,1}$ on m . It is then clear that assumptions made in Sec. III A are too restrictive to provide accurate results for the position of fundamental dipole resonances when m lies between 2.5 and 5. We will therefore attempt a different approach.

Let us keep all the terms in Eqs. (4) and slightly recast them so that the two kinds of Riccati-Bessel functions are separated:

$$\frac{1}{m} \frac{\psi'_1(mx_{\text{res}}^{a_1,1})}{\psi_1(mx_{\text{res}}^{a_1,1})} = \frac{\chi'_1(x_{\text{res}}^{a_1,1})}{\chi_1(x_{\text{res}}^{a_1,1})}. \quad (15)$$

In Fig. 3(a) we present the graphical solution to Eq. (15) for $m = 3.75$. As can be seen, the intersection of $\psi'_1/m\psi_1$ (solid line) and χ'_1/χ_1 (dashed line) takes place for some $x_{\text{res}}^{a_1,1}$ that is located in the vicinity of $x_{\text{res}(1)}^{a_1,1}$, which is the first positive zero of $\psi_1(x, m)$. Therefore, the solution of Eq. (15) is close to an infinite discontinuity (pole) of ψ'_1/ψ_1 . We can then replace such a function by its [0/1] Padé approximant [33] at $x = x_{\text{res}(1)}^{a_1,1}(m)$, that is,

$$\frac{1}{m} \frac{\psi'_1(mx)}{\psi_1(mx)} \approx \frac{1}{m^2(x - x_{\text{res}(1)}^{a_1,1}(m))}. \quad (16)$$

With respect to the right-hand side of Eq. (15), we find it convenient to make use of a [0/1] economized rational approximation (ERA) [34] to $\chi'_1(x)/\chi_1(x)$ over the interval $x_{\text{res}(1)}^{a_1,1}(5) \approx 0.9$ to $x_{\text{res}(1)}^{a_1,1}(2.5) \approx 1.8$ in order to obtain an error distribution that is more uniform than that of the

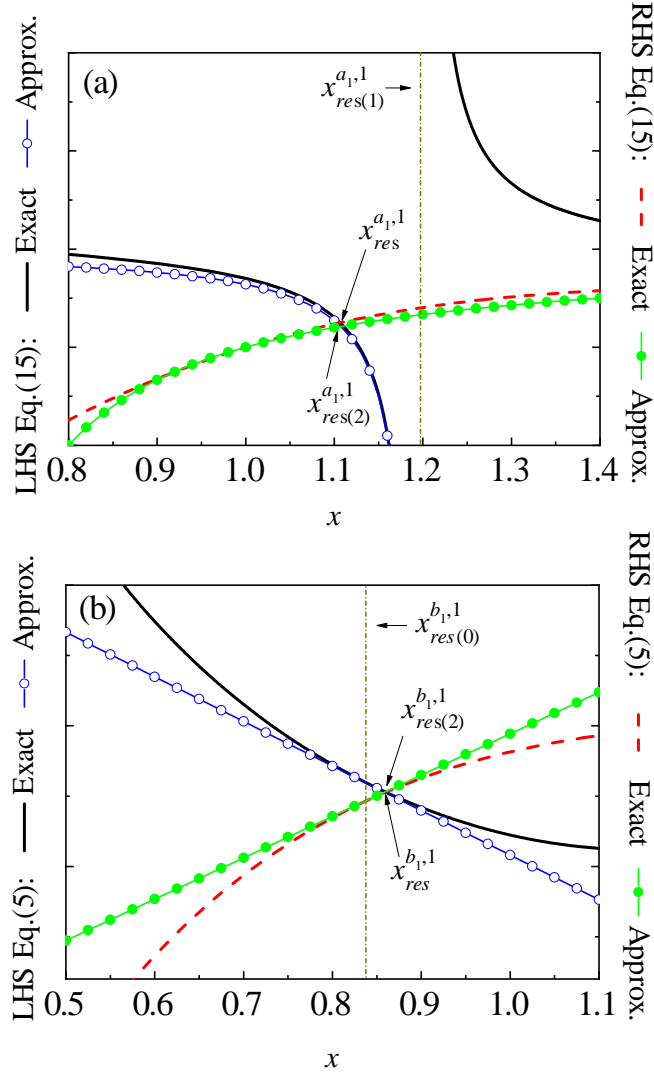


FIG. 3. Graphical determination of the fundamental electric (a) and magnetic (b) dipole resonances for $m = 3.75$. Solid and dashed curves in (a) show the exact values for left- and right-hand sides of Eq. (15), respectively. Open (\circ) and solid (\bullet) symbols denote their proposed approximations (see text). Same conventions apply for Eq. (5) in (b). Vertical dashed-dotted lines mark the positions of $x_{res(1)}^{a1,1}$ and $x_{res(0)}^{b1,1}$.

corresponding Padé approximant about the midpoint:

$$\chi'_1(x)/\chi_1(x) \approx \frac{2}{(3-5x)}. \quad (17)$$

From the right-hand sides of Eqs. (16) and (17) [which are represented in Fig. 3(a) by open and solid symbols, respectively], we finally arrive to an improved explicit expression for the position of the fundamental electric dipole resonance:

$$x_{res(2)}^{a1,1}(m) = \frac{x_{res(1)}^{a1,1}(m) + \frac{3}{2m^2}}{1 + \frac{5}{2m^2}}. \quad (18)$$

For the determination of the fundamental magnetic dipole resonance, we will keep Eq. (5) in its original form. As previously shown in Fig. 2(b), $x_{res}^{b1,1}$ is very close to $x_{res(0)}^{b1,1}$ (in fact, $x_{res}^{b1,1}$ is exactly equal to 1 for $m = \pi$). We can therefore

approximate each side of Eq. (5) by its corresponding linear Taylor expansion about $x = \pi/m$:

$$m\psi'_1(mx)\chi_1(x) \approx L_0(m) + L_1(m)\left(x - \frac{\pi}{m}\right), \quad (19a)$$

$$\chi'_1(x)\psi_1(mx) \approx D_0(m) + D_1(m)\left(x - \frac{\pi}{m}\right), \quad (19b)$$

where

$$L_0(m) = -\frac{m^3}{\pi^3} \cos \frac{\pi}{m} - \frac{m}{\pi} \sin \frac{\pi}{m}, \quad (20a)$$

$$L_1(m) = \left(\frac{m^4}{\pi^4}(4 - \pi^2) - \frac{m}{\pi}\right) \cos \frac{\pi}{m} + \left(\frac{m^3}{\pi^3} + \frac{m^2}{\pi^2}(2 - \pi^2)\right) \sin \frac{\pi}{m}, \quad (20b)$$

$$D_0(m) = \left(1 - \frac{2m^3}{\pi^3}\right) \cos \frac{\pi}{m} - \frac{m^2}{\pi^2} \sin \frac{\pi}{m}, \quad (20c)$$

$$D_1(m) = \left(\frac{8m^4}{\pi^4} - \frac{m^2}{\pi^2} - \frac{m}{\pi}\right) \cos \frac{\pi}{m} + \left(\frac{5m^3}{\pi^3} - 1\right) \sin \frac{\pi}{m}. \quad (20d)$$

The intersection of the right-hand sides of Eqs. (19a) and (19b) provides an excellent approximation to $x_{res}^{b1,1}$, as can be seen in Fig. 3(b) for $m = 3.75$ (open and solid symbols). By expanding $\sin \pi/m$ and $\cos \pi/m$ up to the third order in powers of π/m , the position of the resonance can then be expressed as

$$x_{res(2)}^{b1,1}(m) = x_{res(0)}^{b1,1}(m) + \Delta x_{res}^{b1,1}(m), \quad (21)$$

where $\Delta x_{res}^{b1,1}(m)$ is a quotient of polynomial functions of m . For $m \in [2.5, 5.5]$, we can safely replace it by its [1/2] Padé approximant at $m = \pi$:

$$\Delta x_{res}^{b1,1}(m) \approx \frac{(m - \pi)}{5m^2 - 20m + 35}. \quad (22)$$

Figure 4 shows the calculated values of size parameter as a function of m for the fundamental electric and magnetic dipole resonances of a nonabsorbing dielectric sphere with its relative refractive index between 2.5 and 5. Open symbols (\circ) denote the numerical solutions to Eqs. (4) and (5), whereas solid, dashed, and dotted lines show the values of the proposed approximations to $x_{res}^{a1,1}$ and $x_{res}^{b1,1}$ with subscripts (0), (1), and (2), respectively. As can be seen in Fig. 4(a), the size parameter corresponding to the fundamental electric dipole resonance ranges between 1.45 for $m = 2.5$ and 0.87 for $m = 5$. These values are systematically overestimated by $x_{res(0)}^{a1,1}$, which bears a percentage error that increases from $\%_{error} = +8$ for $m = 5$ to $\%_{error} = +28$ for $m = 2.5$. As regards $x_{res(1)}^{a1,1}$, one may observe that it shows an acceptable agreement with the numerical results about $m = 5$ and then becomes much less reliable as m decreases ($\%_{error} = +21$ for $m = 2.5$). On the other hand, proposed $x_{res(2)}^{a1,1}$ keeps the absolute value of percentage error below 2 for the entire range of refractive index values, thus providing a significant improvement to the approximate determination of $x_{res}^{a1,1}$.

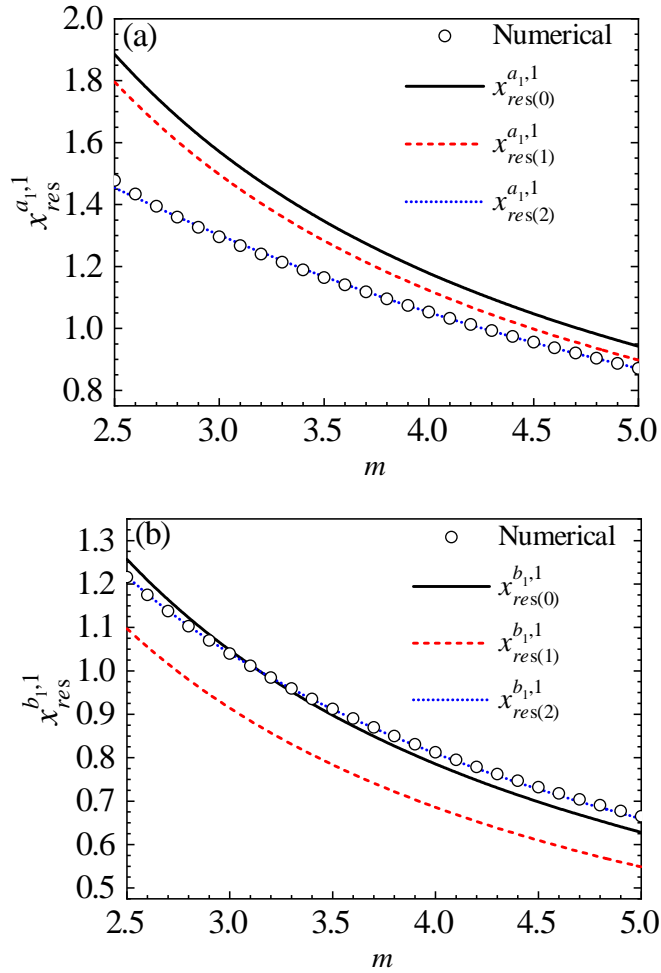


FIG. 4. Calculated size parameter as a function of m for the fundamental electric (a) and magnetic (b) dipole resonances. Open symbols (\circ) in (a) correspond to the numerical solution to Eq. (4), whereas solid, dashed, and dotted curves show the values of the proposed approximations to $x_{res}^{a1,1}$ with subscripts (0), (1), and (2) (see text). Same conventions apply for Eq. (5) and $x_{res}^{b1,1}$ in (b).

With respect to the fundamental magnetic dipole resonance, we have already mentioned that $x_{res(0)}^{b1,1}$ provides an estimate for $x_{res}^{b1,1}$ that is exact for $m = \pi$. As shown in Fig. 4(b), the size parameter is slightly overestimated for m below π , whereas it is equivalently underestimated for m above such threshold. The absolute value of percentage error does not exceed 6 for any considered value of m . Unlike $x_{res(1)}^{a1,1}$, $x_{res(1)}^{b1,1}$ does not improve the approximation to the resonance. Conversely, it consistently underestimates the resonant size parameter throughout the interval with a percentage error that ranges between $\%_{error} = -10$ for $m = 2.5$ and $\%_{error} = -18$ for $m = 5$. Fortunately, the $x_{res(2)}^{b1,1}$ approximation is in turn found to be 99% accurate for the range between $m = 2.5$ and 5, which shows the convenience of this approach.

In order to better understand the very different reliability of approximations with subscript (0) for the fundamental electric and magnetic dipole resonances, let us now consider an issue that seems at first sight unrelated to the subject, namely, the determination of fundamental dipole antiresonances. If either $|a_1|^2$ or $|b_1|^2$ vanish for $m > 1$, dipole contributions

to scattering are then suppressed, thus producing a noticeable deep in Q_{sca} (see, e.g., Fig. 1) unless other multipolar orders be dominant. Dipole resonances and their antagonists are, by the way, very close to each other so that plots of $|a_1|^2$ and $|b_1|^2$ as a function of either x or m exhibit a Fano-type line shape [23,35,36]. Aside from their fundamental interest, dipole antiresonances may also be relevant by themselves for the designing of dielectric nanoresonators [37].

According to Eq. (2), an electric dipole antiresonance is expected to happen whenever $p_1(x, m)$ is equal to zero. By following exactly the same procedure as in Sec. III A, we obtain that approximations with subscripts (0) and (1) for the fundamental electric dipole antiresonance do coincide with those of $x_{res}^{a1,1}$:

$$x_{antires(0)}^{a1,1}(m) = x_{res(0)}^{a1,1}(m) = \frac{3\pi}{2m}, \quad (23)$$

$$x_{antires(1)}^{a1,1}(m) = x_{res(1)}^{a1,1}(m) = \frac{3\pi(9\pi^2 - 8)}{2m(9\pi^2 - 4)}. \quad (24)$$

It is not but up to approximation with subscript (2) [that is, [0/1] Padé for $\psi'_1(mx)/m\psi_1(mx)$ and [0/1] ERA for $\psi'_1(x)/\psi_1(x)$] that size parameters of resonance and antiresonance depart from each other:

$$x_{antires(2)}^{a1,1}(m) = \frac{x_{antires(1)}^{a1,1}(m) - \frac{5}{7m^2}}{1 - \frac{8}{7m^2}}. \quad (25)$$

In contrast, approximation with subscript (0) for the fundamental magnetic dipole antiresonance leads us to

$$x_{antires(0)}^{b1,1}(m) = \frac{2\pi}{m} \quad (26)$$

and not to π/m . The subsequent linear expansion of

$$r_1(x, m) = 0 \quad (27)$$

about $x_{antires(0)}^{b1,1}$ allows one to obtain

$$x_{antires(2)}^{b1,1}(m) = x_{antires(0)}^{b1,1}(m) + \Delta x_{antires}^{b1,1}(m), \quad (28)$$

with

$$\Delta x_{antires}^{b1,1}(m) \approx \frac{58(99 - 46m)}{5(1100m^2 - 2155m + 425)}. \quad (29)$$

Let us now revisit the scattering response of a dielectric sphere with $m = 3.75$ in the guise of the squared norms of a_1 and b_1 . Their corresponding line shapes in Fig. 5(a) show very close maxima and minima and, in particular that for b_1 , are definitely Fano type. As can be seen, the position of fundamental antiresonances (either electric or magnetic) agrees very well with approximations with subscript (2) that are marked with vertical dashed-dotted lines. Interestingly enough, it is also apparent that approximation with subscript (0) is in fact much closer to the fundamental electric dipole antiresonance than to its resonant counterpart, unlike that for the magnetic dipole one (vertical solid lines). As shown in Fig. 5(b), there is a good agreement between $x_{res/antires(0)}^{a1,1}$ and $x_{antires(2)}^{a1,1}$ for all considered values of m , which closely resembles that between $x_{res(0)}^{b1,1}$ and $x_{res(2)}^{b1,1}$ in Fig. 4(b). We can therefore end our discussion with the conclusion that solution of Eq. (8), although needed in order to finally obtain an

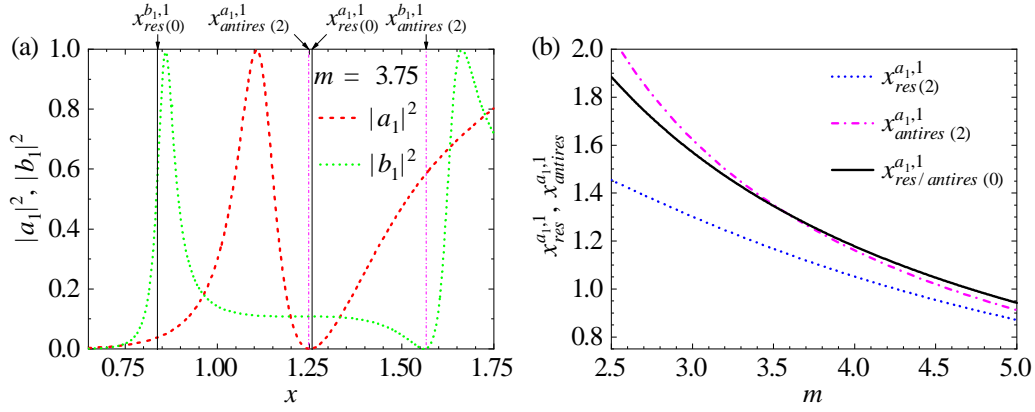


FIG. 5. (a) Calculated values of $|a_1|^2$ (dashed) and $|b_1|^2$ (dotted) as a function of size parameter x for a dielectric sphere with $m = 3.75$. Vertical solid lines show the approximated values of fundamental dipole resonances with subscript (0), whereas dashed-dotted ones mark the positions of approximated antiresonances with subscript (2). (b) Calculated size parameters as a function of m for the approximations with subscript (2) to the fundamental electric dipole resonance (dotted) and antiresonance (dashed-dotted). Solid curve shows the values of proposed approximations with subscript (0).

accurate approximation to $x_{\text{res}}^{a_1,1}$, does in fact provide by itself a good estimate for $x_{\text{antires}}^{a_1,1}$ rather than for the position of the fundamental electric dipole resonance.

C. Approximations to $x_{\text{res}}^{a_1,j}$ and $x_{\text{res}}^{b_1,j}$ for $j > 1, mx \gg 1$

When considering dipole resonances with different ordinal number (that is, for $j > 1$), the solutions to Eqs. (4) and (5) will no longer be close to $x_{\text{res}} = 1$, but may take much larger values. This implies that the condition $mx \gg 1$ can then be met for some values of m that are significantly smaller than those considered in Sec. III A. In such a scenario, it seems again plausible to disregard nonsinusoidal terms in Eqs. (6a) and (6b) but one can not take for granted the validity of Eqs. (7). Consequently, we should keep contributions from $\chi_1 \psi_1'$ and $\chi_1' \psi_1$ when defining the approximations to q_1 and s_1 for $x \gg 1$:

$$q_1^{x \gg 1}(x, m) = m \cos x \cos mx + \sin x \sin mx, \quad (30a)$$

$$s_1^{x \gg 1}(x, m) = -m \sin x \sin mx - \cos x \cos mx. \quad (30b)$$

As can be seen in Fig. 6, zeros of $q_1^{x \gg 1}$ agree with those of q_1 for $x \gtrsim 3$ over the entire range of considered refractive index values. For the case of s_1 and $s_1^{x \gg 1}$, such an agreement can be found for $x \gtrsim 4$. Hence, the positive solutions to

$$m \cos x \cos mx + \sin x \sin mx = 0, \quad (31a)$$

$$m \sin x \sin mx + \cos x \cos mx = 0 \quad (31b)$$

will provide a good approximation to successive electric and magnetic dipole resonances with $j > 1$, respectively.

In order to obtain those solutions, we now return to our previous discussion of $x_{\text{res}}^{a_1,j}$ and $x_{\text{res}}^{b_1,j}$. For the case of electric dipole resonances, let $j > 1$ and $m \gg 1$, so that the position of resonances is governed by the condition $m \cos x \cos mx = 0$. Hence,

$$x_{\text{res}}^{a_1,j>1}(m) \approx x_{\text{res}^{(0)}}^{a_1,j>1}(m) = \frac{(j + \frac{1}{2})\pi}{m}. \quad (32)$$

As m goes down, $x_{\text{res}}^{a_1,j>1}$ should go up inversely, due to the continuity of size parameter and its inverse dependence on m . But, such a continuous increase also implies that

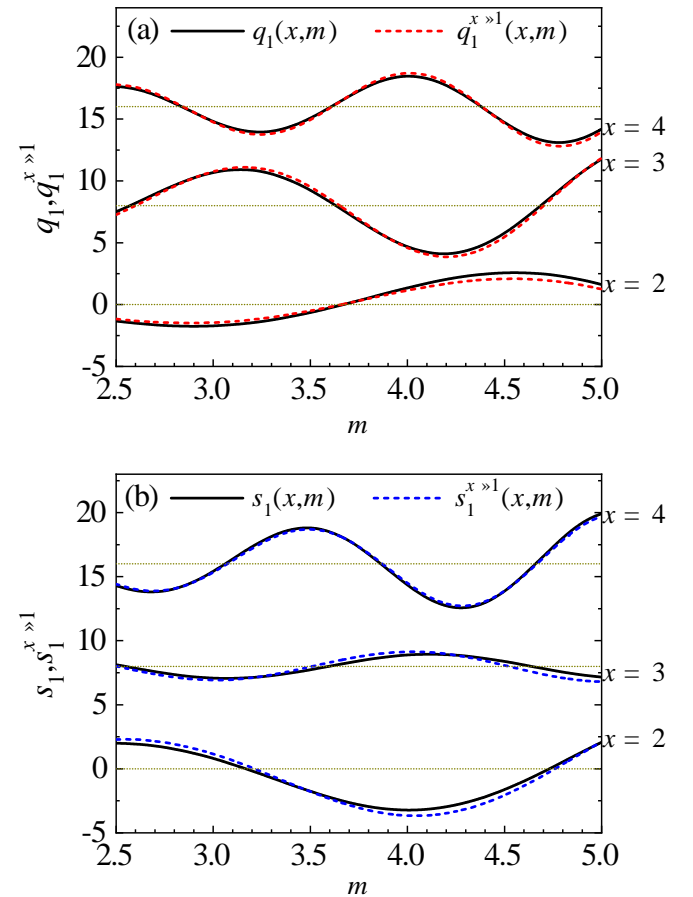


FIG. 6. (a) Calculated $q_1(x, m)$ (solid) and $q_1^{x \gg 1}(x, m)$ (dashed) as a function of m for increasing values of size parameter. Curves corresponding to $x = 2, 3, 4$ are artificially shifted for the sake of clarity, with their actual zero baselines marked by horizontal dotted lines. (b) Same conventions apply for $s_1(x, m)$ and $s_1^{x \gg 1}(x, m)$.

resonant x should equal $(g + \frac{1}{2})\pi$ for some given m , with $g = 1, 2, 3, \dots$. According to Eq. (32), we expect it to happen for $m = (j + \frac{1}{2})/(g + \frac{1}{2})$, but the couplet

$$(x, m) = \left((g + \frac{1}{2})\pi, \frac{(j + \frac{1}{2})}{(g + \frac{1}{2})} \right) \quad (33)$$

is not a solution of Eq. (31a), which is reduced to $\sin m(g + \frac{1}{2})\pi = 0$ for $x = (g + \frac{1}{2})\pi$. In fact, it is

$$m_g^{a1,j} = \frac{(j + g)}{(g + \frac{1}{2})} \quad (34)$$

that fulfills Eq. (31a) for that particular x . A not-so-obvious consequence of this mathematical condition is that every time when m equals $m_g^{a1,j}$, the resonant size parameter experiences a “jump” of $\frac{\pi}{m}$ opposite to the variation in m , then promoting or demoting to the adjacent zeroth-order resonance.

We can then expect the position of electric dipole resonances with $j = 2, 3, \dots$ to be approximately described by

$$x_{\text{res}(3)}^{a1,j}(m) = x_{\text{res}(0)}^{a1,j}(m) + \frac{\pi}{m} \sum_{g=1}^{\infty} [1 - H(m - m_g^{a1,j})], \quad (35)$$

where $H(m)$ is a smooth analytical approximation to the Heaviside step function [38]

$$H(m) = \frac{1}{2} + \frac{1}{\pi} \arctan hm \quad (36)$$

in which h is left as a free parameter.

Following the same reasoning for magnetic dipole resonances, we obtain

$$x_{\text{res}(3)}^{b1,j}(m) = x_{\text{res}(0)}^{b1,j}(m) + \frac{\pi}{m} \sum_{g=1}^{\infty} [1 - H(m - m_g^{b1,j})], \quad (37)$$

with

$$m_g^{b1,j} = \frac{(j + g - \frac{1}{2})}{g} \left(1 + \frac{\delta_{g1}}{\pi^3 - 2} \left[1 - \frac{3}{(j + \frac{1}{2})^2} \right] \right), \quad (38)$$

where δ_{g1} is the Kronecker delta [39]. Please notice that the extra summand for $g = 1$ is due to the discrepancy between the zeros of $s_1(\pi, m)$ and those of $s_1^{\gg 1}(\pi, m)$ (see Appendix).

Figure 7 shows the calculated values of size parameter as a function of m for successive electric and magnetic dipole resonances with $j > 1$. Solid lines correspond to numerical solutions of Eqs. (4) and (5), whereas dashed ones represent the best fits of expressions in Eqs. (35) and (37) to data points. Free parameter h is determined for every (j, g) by means of an iterative implementation of the Levenberg-Marquardt algorithm [40,41]. For a given g , we find h to be negatively proportional to j . On the other hand, h is directly proportional to g if j is kept as a constant.¹ As can be seen, expressions

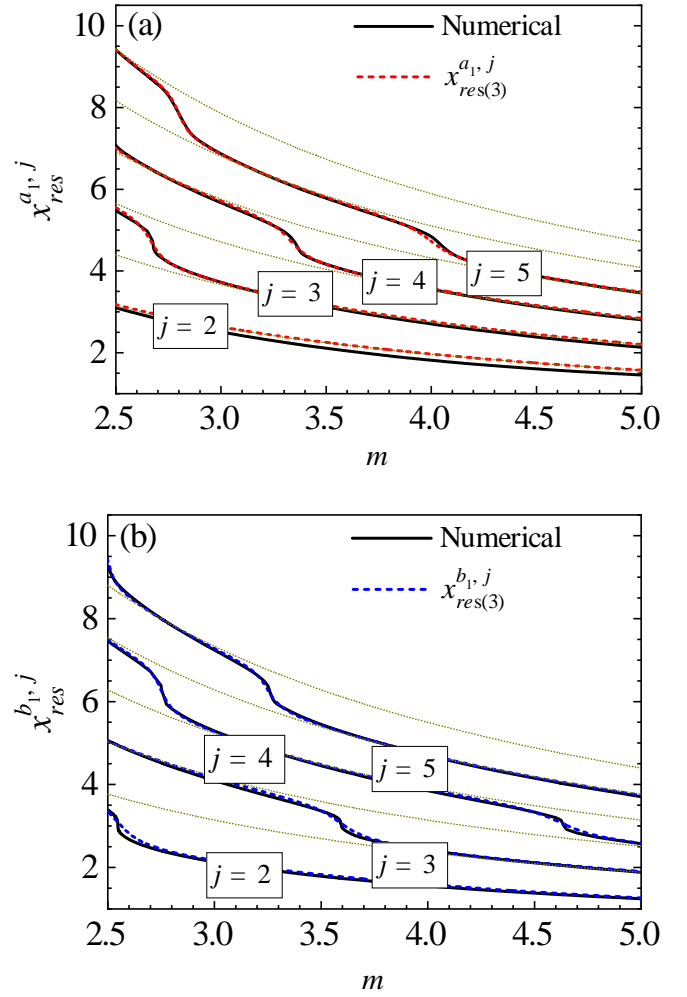


FIG. 7. Calculated size parameter as a function of m for higher-order electric (a) and magnetic (b) dipole resonances of a nonabsorbing dielectric sphere. Solid curves in (a) correspond to the numerical solutions of Eq. (4) for increasing values of j , whereas dashed curves show the best fits of our proposed $x_{\text{res}(3)}^{a1,j}$ to data points (see text). Dotted curves mark the positions of sequential zeroth-order resonances. Same conventions apply for Eq. (5) and $x_{\text{res}(3)}^{b1,j}$ in (b).

with subscript (3) provide a reliable description of dipole resonances with $j > 1$ for the range between $m = 2.5$ and 5, especially with respect to “jumps” between adjacent zeroth-order solutions. In addition, curve fitting to data points keeps the absolute percentage error below 2.5 all over the considered refractive index range.²

From Fig. 7 it is also apparent that electric and magnetic dipole resonances with $j > 1$ do coincide for precise values of the relative refractive index (e.g. $m \approx 2.55$ for $j = 2$). However, the occurrence of “double-dipole” resonances does not seem to cause any significant effects due to the dominance of contributions other than dipole.

¹Obtained values for h in Fig. 7 can be approximated by $(3g - 1)(14.52 - 1.82j)$ and $(2g - 1)(15.17 - 1.66j)$ for the case of electric and magnetic dipole resonances, respectively.

²With the sole exception of $x_{\text{res}(3)}^{a1,2}$, which systematically overestimates $x_{\text{res}}^{a1,2}$ for $m > 2$. Percentage error reaches its maximum (+10%) at $m = 4.5$.

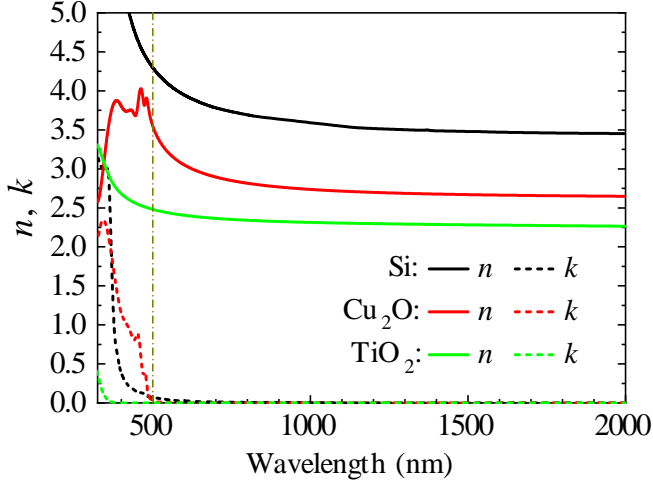


FIG. 8. Real (solid) and imaginary (dashed) parts of the refractive index as a function of wavelength for Si, Cu₂O, and TiO₂, obtained from Refs. [32,42,43], respectively. Vertical dashed-dotted line marks the wavelength from which absorption can be neglected.

IV. DIPOLE RESONANCES FOR HIGH- AND MODERATE-REFRACTIVE-INDEX MATERIALS

Up to this point, we have been focused on the solution of equations. We now return to the scattering properties of actual dielectric nanospheres in the optical range. For the sake of simplicity, let us assume that our sphere is surrounded by air, so that we can replace m by the sphere's complex refractive index $n + ik$. Given that all our findings have been obtained for nonabsorbing materials, we require $k \approx 0$. In Fig. 8 we present the real and imaginary parts of the refractive index as a function of wavelength for Si, Cu₂O, and TiO₂, according to Refs. [32,42,43], respectively. As can be seen, these three materials fulfill the requirement of not absorbing light within the interval between 500 and 2000 nm and have therefore been the subject of recent experimental research on dielectric nanoresonators [21,22,44–49]. In addition, the range of values of n within such interval for Si, Cu₂O, and TiO₂ cover most of that of m that was considered in the previous section. These materials seem, then, to be convenient to test our improved approximate conditions by comparing their predicted resonances with those obtained from the full calculation of $Q_{\text{sca}}^{a_1}$ and $Q_{\text{sca}}^{b_1}$.

It follows from the very definition of size parameter that

$$R_{\text{res}}^{\square,1} = \frac{\lambda}{2\pi} x_{\text{res}}^{\square,1}(m(\lambda)), \quad (39)$$

where \square stands for either a_1 or b_1 . We then substitute for $x_{\text{res}}^{\square,1}$ from Eqs. (18) and (21) into Eq. (39) in order to obtain the best estimates for (m, λ, R) triplets exhibiting fundamental dipole resonances. For the sake of comparison, we also calculate the resonant radii corresponding to approximations with subscript (0).

Panels in Fig. 9 show the calculated electric and magnetic dipole contributions to the scattering efficiency of a dielectric sphere as a function of the incident wavelength and the sphere's radius for Si, Cu₂O, and TiO₂. All calculated values result from straightforward evaluation of $Q_{\text{sca}}^{a_1}$ and $Q_{\text{sca}}^{b_1}$ by

means of a homemade *Mathematica* script.³ Open (\circ) and solid (\bullet) symbols correspond to the above-mentioned $R_{\text{res}(0)}^{\square,1}$ and $R_{\text{res}(2)}^{\square,1}$, respectively. As can be seen, there is an excellent agreement between the latter and numerical results for all three materials across the considered wavelength range. In addition, it is apparent that, by increasing the wavelength, estimates with subscript (0) depart from the actual resonances exactly in the same fashion as they do $x_{\text{res}(0)}^{a_1,1}$ and $x_{\text{res}(0)}^{b_1,1}$ when m decreases (see Fig. 4).

In fact, Fig. 9 brings us up against some physical interpretation of resonant (m, λ, R) triplets. From considerations based on geometrical optics (see, for example, Ref. [24]), it can be figured out that electric dipole resonances occur when $2R$ becomes approximately equal to an odd multiple of the half-wavelength inside the sphere, which is precisely the prediction of Eq. (11). However, Figs. 9(a), 9(c), and 9(e) show that, as m decreases, electric dipole resonances take place for radii that are smaller than $R_{\text{res}(0)}^{a_1,1}$, thus pointing out some sort of effective increasing of the sphere's size for moderate values of m . In contrast, there is no such resizing for magnetic dipole resonances, which appear for diameters that are equal to an integer multiple of λ/m , aside from the correction prescribed by Eq. (22).

With respect to such correction, we can notice the threshold at $m = \pi$ when comparing the three different materials: as the refractive index of silicon is always higher than π , the values of $R_{\text{res}(0)}^{b_1,1}$ are consistently lower than those of $R_{\text{res}(2)}^{b_1,1}$, whereas it becomes just the opposite for TiO₂. On the other hand, the refractive index of Cu₂O equals π about 570 nm, so that the crossing between the two estimates is hard to ascertain from Fig. 9(c). From such wavelength on, cuprous oxide follows the same trend as titanium dioxide. Finally, signatures of resonances with $j = 2$ are clearly apparent in the upper left quadrant of every panel in Fig. 9. Nevertheless, their corresponding scattering efficiencies are about one fifth of those of the fundamental resonances and they do not seem to be especially relevant for any of these materials.

Given that the zero-absorption threshold defined in Fig. 8 is somewhat arbitrary, we cannot close this section without discussing the robustness of our obtained approximations when dealing with some degree of dissipation. For a complex relative refractive index $m = m' + im''$, there is no unequivocal correspondence between resonances and antiresonances in $Q_{\text{sca}}^{a_1}$ and $Q_{\text{sca}}^{b_1}$ and zeros and poles in a_1 and b_1 , so that explicit expressions for resonant or antiresonant values of size parameter become practically unattainable. However, one could expect approximations based on the real part of m to still hold for a weakly absorbing medium.

As a test for this hypothesis, we present in Fig. 10 the calculated values of electric and magnetic dipole contributions to the scattering efficiency as a function of size parameter x for a dielectric sphere with $m = 3.75 + im''$. Such a fixed value for m' corresponds to the midpoint of the interval between 2.5 and 5 that has been considered all along this work. It is also in the range between those of n for Si and Cu₂O at the

³Wolfram Research, Inc., Mathematica, Version 9.0, Champaign, IL (2012).

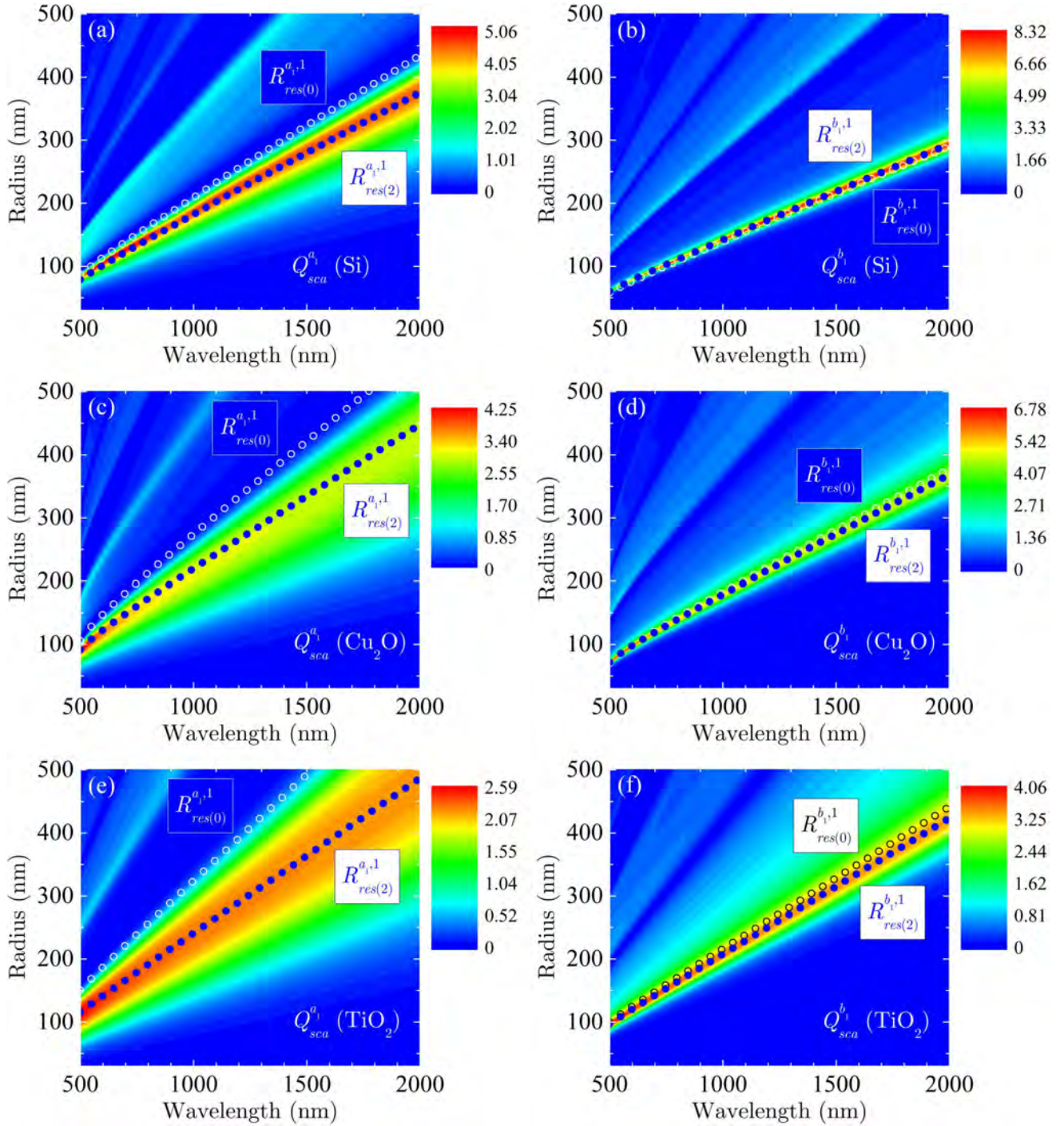


FIG. 9. Electric and magnetic dipole contributions to the scattering efficiency of a dielectric sphere as a function of the incident wavelength and the sphere's radius for three different materials: Si (a), (b), Cu_2O (c), (d), and TiO_2 (e), (f). Open (\circ) and solid (\bullet) symbols correspond to estimates for resonant radii with subscripts (0) and (2), respectively.

wavelength from which absorption seems negligible in Fig. 8. With respect to m'' , it is gradually increased from 0 to 1, which is the maximum value of κ for silicon and cuprous oxide above 400 nm. Please keep in mind that we have chosen this setting for testing purposes only, as far as m'' is connected with m' by causality and cannot therefore take arbitrary values.

As can be seen, all spectral features are significantly damped and also slightly shifted as dissipation increases.

Direction of the spectral shift with respect to approximations with subscript (2) for $m = 3.75$ (vertical dashed lines) seems to be opposite for fundamental electric and magnetic resonances and antiresonances. Thus, the position of fundamental electric dipole resonance shifts from $x = 1.10$ for $m'' = 0$ to $x = 1.06$ (-4%) for $m'' = 0.5$ and then returns to $x = 1.09$ for $m'' = 0.75$, which is the highest of the considered values that permits the resolution of the peak. In

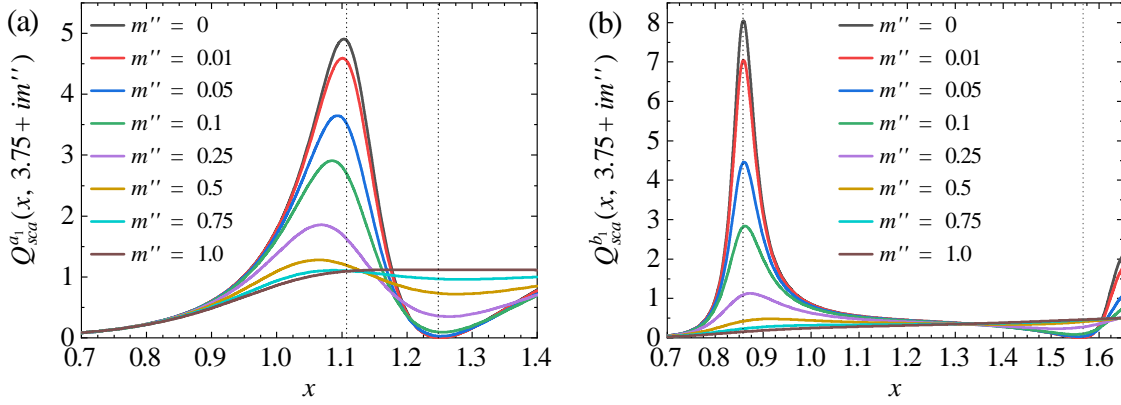


FIG. 10. Electric (a) and magnetic (b) dipole contributions to the scattering efficiency as a function of size parameter x for a dielectric sphere with $m = 3.75 + im''$. Solid curves correspond to increasing values of m'' . Vertical dashed lines mark the approximated positions of fundamental dipole resonances and antiresonances for $m = 3.75$.

contrast, $x_{\text{antires}}^{a_1,1}$ shifts oppositely (+1%) for $m'' = 0.75$. On the other hand, the size parameter of fundamental magnetic dipole resonance evolves from $x = 0.86$ for $m'' = 0$ to $x = 0.91$ (+6%) for $m'' = 0.5$, whereas that for the antiresonance does from $x = 1.56$ to $x = 1.41$ (−10%). We can then conclude that Eqs. (18), (21), (25), and (28) can be reasonably extended to the entire visible range for Si, Cu₂O, and TiO₂, which seems convenient for designing purposes.

V. CONCLUSIONS

We have obtained explicit expressions that provide accurate approximations to dipole resonances and antiresonances in the scattering spectrum of nonabsorbing dielectric nanospheres with high- and moderate-refractive-index values in the optical range. These expressions enable us to predict the occurrence of a dipole resonance with any ordinal number for a triplet of sphere's radius R , incident wavelength λ , and relative refractive index value m without the actual evaluation of Mie scattering coefficients. Our predictions retrieve previous results for $m \gg 1$ and extend them to a wider range. We have confirmed their validity for specific dielectric materials that are widely used in photonic devices. Therefore, we expect our results to be useful for the designing of dielectric nanoresonators, particularly for issues such as biosensing [48], nanoscopy [50], or photonic nanojet lithography [51].

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APPENDIX: DETERMINATION OF $m_1^{b_1,j}$

For $x \gg 1$, we expect the solutions of $s_1^{x \gg 1}(x, m) = 0$ to coincide with those of $s_1(x, m) = 0$. However, as can be seen

in Fig. 11, there is still some discrepancy between them for $x = \pi$ (i.e., $g = 1$). In order to ensure both their accuracy and their closeness, we have defined $m_1^{b_1,j}$ as the zeros of the linear Taylor expansion of $s_1(\pi, m)$ about $m = (j + \frac{1}{2})$, which can be expressed as

$$m_1^{b_1,j} = \left(j + \frac{1}{2}\right) \left(1 + \frac{1}{\pi^3 - 2} \left[1 - \frac{3}{(j + \frac{1}{2})^2}\right]\right). \quad (\text{A1})$$

This leads to zeros that are slightly greater than $(j + \frac{1}{2})$:

$$m_1^{b_1,\{2,3,4,\dots\}} = \{2.54482, 3.59111, 4.63216, \dots\}. \quad (\text{A2})$$

By the restriction of this correction to $g = 1$, we finally arrive to Eq. (38).

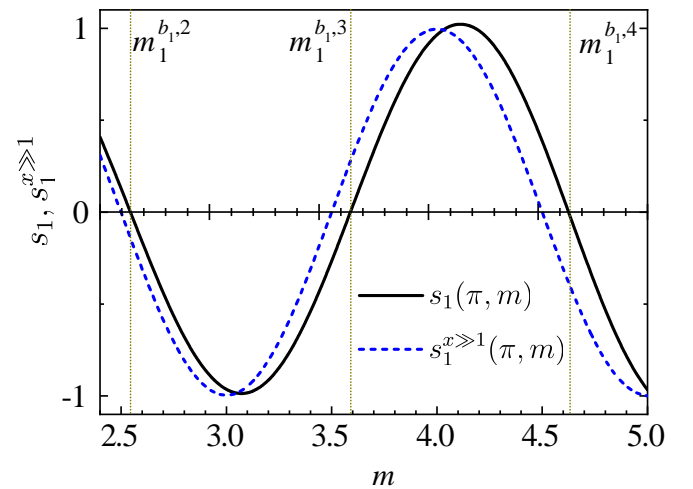


FIG. 11. Calculated $s_1(\pi, m)$ (solid) and $s_1^{x \gg 1}(\pi, m)$ (dashed) as a function of m . Vertical dashed-dotted lines mark the values of $m_1^{b_1,j}$ from Eq. (A2).

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